Radionuclide Distribution and Transport in Terrestrial and Aquatic Ecosystems

A Critical Review of Data

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VOLUME ONE

Prepared for the Directorate-General Employment, Social Affairs and Education, Health and Safety Directorate, Commission of the European Communities

A.A.BALKEMA / ROTTERDAM / 1983

2. Models for soils and plants.

A simplified soil-plant model is represented in Fig. 2.1. Soil can be considered in terms of four components; an organic component, a mineral component, a parent material component and the soil solution. This separation is somewhat artificial, since, in practice, it is often difficult to separate organic, mineral and solution phases. Moreover, parent-material cannot be truly considered part of the soil system, since it is the source of derivation of soil. Thus, in environmental studies of radionuclides after atmospheric or aquatic releases, the transfer from parent material to mineral soil is irrelevant.

Within any component there are several sub-divisions. For example, it should be possible to isolate a surface organic component, a mixed organic component, and both a dissolved and suspended organic component within the soil organic fraction. In many studies soil has been considered simply as a series of layers of increasing depth. Such a representation takes no account of the differences in physical and chemical properties between the different horizons of any soil profile, or, in a disturbed profile, between organic and mineral phases. Furthermore, plants are often considered in isolation from soil with insufficient regard for their root component. This latter component is often difficult to distinguish from soil components, but is of considerable importance in the uptake, binding and loss of trace substances by the plant.

Input to the various soil components occurs via wet and dry deposition, and via artificial application (e.g. irrigation). The degree of surface deposition depends upon a variety of factors other than the concentration of activity in air. Retention within the surface material depends upon:

- the physical and chemical nature of the input;
- the degree of resuspension of the surface layer;
- the degree of biological decomposition occurring;
- the degree of leaching of the soil surface.

It can be assumed that the organic component is in close contact with the soil mineral lattice and receives an input from this component via leaching and biological activity. Leaching may carry material from the soil through the parent material and away from the system via percolation into ground water.

In Fig. 2.1 the three main solid-phase components are shown exchanging with the soil solution. This solution component is included as an intermediate between any of the solid components and the plant-root component. It is assumed that all materials for plant uptake pass through this component, as do materials released by root mortality and subsequent biological decay. A root component is included in the model, since this represents an important method of contaminant return to soil after ploughing, allows for more reliable estimation of translocation and is relevant in any assessment of the impact of harvesting of root and/or tuber crops for human consumption. Within the plant, radionuclides may be either translocated from root to shoot (as in most cereal crops) or transported from shoot to root for storage (as in most root vegetable and tuber crops).

The above-ground parts of the plant can be considered as two main components; the external component which receives an aerial input via deposition of particulate matter from the atmosphere and translocated materials from the rest of the plant; and an internal component which mediates in absorption, translocation and storage processes. Both compon-

10 in pasture plants by seasonal mortality and by abscis- rts which results in a return to the soil. The roots, or the whole plant may be harvested for human and/or . After harvesting, the above-ground external component ses, be lost or removed to a large extent (by mechanical ocesses) prior to human consumption. A fruiting component is ed in this general model (Fig. 2.1), mainly because of its sea- ature, however in more detailed studies of individual elements component is considered where data are available.

2.1 Soils

2.1.1 Radionuclide input to soils

Inputs to soil occur from the processes of wet and dry deposition, irrigation, animal excretion, sewage sludge application, application of fertilizers and the decay of senescent, cut and dead plant or animal materials.

Dry deposition is the process by which particulates are transferred from the atmosphere to the soil surface and the total extent of deposition is estimated using the concept of deposition velocity (V_g). In previous studies [e.g. 15] a value of 5×10^{-3} m s^{-1} has been adopted for this parameter to represent particulate matter of several microns diameter. However, it has been noted that V_g is variable over several orders of magnitude, depending upon particle size, the reactivity of the aerosol, the physical conditions of the soil surface, and the meteorological conditions at the time of deposition. The value of 5×10^{-3} m s^{-1} was reported to have been adopted on the basis of the data of Slinn [70]. Reference to the discussion of Slinn does not necessarily accord with the proposed value for V_g. Slinn reviewed available data for V_g and stated in conclusion that "the values of V_w (wet deposition velocity) and V_g, typically about 1×10^{-2} to 5×10^{-2} m s^{-1}, should be within a factor of 5 of the true annual-average values and may be within a factor of 2". During the derivation of working limits for the release of radionuclides to agricultural systems, Bryant [12] noted Chamberlain's calculations for V_g of 10^{-3} to 3×10^{-3} m s^{-1} for submicron particles and 10^{-2} m sec^{-1} for 15 μm diameter particles. A review of the published data for V_g reveals very few measured values for soil surfaces as opposed to plant surfaces. However, the data that are available suggest that the assumption that vegetation and soil surfaces are similar in their ability to collect airborne material is suspect. Little [47] has reported, in a summary of his experimental data, deposition velocities for exhaust lead particles (0.2 μm diameter) to soil of 3.6×10^{-5} to 5.3×10^{-4} m s^{-1}, these were considerably lower than those reported for vegetative surfaces in comparable studies.

In the context of agricultural systems, the surface for collection of aerosols is unlikely to be either purely vegetative or purely soil. It is most likely to be a combination of both types. Little (op. cit.) referred to measurements of V_g for 2.75 and 5.0 μm diameter polystyrene particles depositing onto both grass and soil surfaces. In both cases the large majority of the total catch was associated with the grass and only 10 to

35% was deposited directly to the soil under the grass. In the case of exhaust aerosols, the presence of grass 10 cm in height increased total deposition to the same area by a factor of 3 to 4 times. Little proposed that the majority of the catch was where turbulence was greatest around the top of the sward, the rapid decline in wind speed towards the soil surface inhibited mixing and reduced deposition rates close to the surface.

V_g is markedly affected by wind speed and the median aerodynamic diameter of the particles concerned. Although the commonly quoted value of 5×10^{-3} m s^{-1} is suggested to be typical of particles of an aerodynamic diameter of several microns depositing onto a wide variety of surfaces, reference to the data of Slinn [70] and Little [47] suggests that this value is in fact representative only of particles less than about 3 μm diameter. Deposition velocities for larger particulates on all surfaces can be expected to be considerably greater than this value, but, in general, the V_g for soil can be expected to be considerably less than that for vegetation under similar conditions. Further data concerning deposition velocities are discussed in Section 2.2.1.

Like dry deposition, the extent of wet deposition has usually been estimated by the use of a simple coefficient, the washout coefficient (Λ). Generally speaking, the procedure for estimating wet deposition involves the assumption that rainfall is continuous throughout the transit of a particular fraction of a hypothetical plume. The wet deposition approach to simulation modelling of radionuclide transfer has been discussed by Slinn [70] who noted that washout rates could vary by more than an order of magnitude during a single rainstorm and that washout was not necessarily directly proportional to rainfall rate. Ritchie et al. [62] noted that the theoretical calculation and experimental measurement of Λ are fraught with difficulty, mostly because the extent of dry deposition occurring with wet deposition is not known; other complications included drop evaporation, turbulence and electrical effects. Rainfall is only considered in specific Pasquill and Doury categories and in these conditions a value of $\Lambda = 10^{-4}$ s^{-1} was suggested by the CEA/NRPB [15] to be appropriate to small particulates and a rainfall rate of a few millimetres per hour (typical of Western Europe). It should be noted that the choice of values for Λ vary between authors, such that Bayer [4] in similar studies to those of the CEA/NRPB [15] made use of a value of 2.8×10^{-5} s^{-1} for the Rhine-Meuse region.

At the time of writing, data concerning parameters used to estimate the input of a contaminant into soil via wet and dry deposition processes appear to be rather limited and extremely variable. Furthermore, very few of the extensive studies of deposition processes have limited their considerations to soil surfaces as opposed to vegetative surfaces.

Inputs to soil via irrigation, animal excretions, sewage sludges, application of fertilizers and from decaying or dead organic materials are generally not considered in assessment studies. These inputs can be considered in most cases to be of a secondary nature and their estimation should be relatively easy given site-specific parameters. However, in the case of radionuclides with radioactive half-lives in excess of 100 days, these pathways could become significant in the longer-term distribution of a deposited radionuclide in the soil ecosystem. In this context, it is relevant to note that cows can ingest up to 450 kg of soil per year and sheep up to 23 kg per year with their normal diet [36]. To a large extent, this will be surface soil and the fraction of a radionuclide that is not absorbed by the animal will be returned to the pasture. In the case of non-permanent agricultural pastures or crops, the portion of the biomass that is not harvested will often be returned to the soil system at the end of the season, and this is especially true of the root component. In permanent pastures, a considerable proportion of the radionuclide will be returned to soil, either at the end of the growing season as a result of die-back, or during the season due to either die-back or abscission/dehiscion of plant parts. The extent of this input will be related to the form of the pasture and its species composition (as well as to meteorological conditions throughout the growing season). In this context, it should be noted that there is a considerable body of relevant information concerning the processes of straw decay in field conditions [e.g. 33].

Decomposition is an important process in both terrestrial and aquatic ecosystems and an introduction to the subject is given by Mason [49]. Detritus particles which are derived from decomposition of vascular plants have a high sorptive capacity for a wide range of pollutants and the importance of studying detritus as a standard component in pollutant release assessments has been stressed [59]. Turnover times for calcium in litters of various forests range from 0.2 to 35 years and Jordan and Kline [42] stated that these cycling times were influenced primarily by the rates of element uptake and release by plants. There are few data for the rate of release of radionuclides from litters [e.g. 80], but experimental studies show that a high proportion of added-radionuclides can be sorbed rapidly by litter [e.g. 41].

The application of sludges and fertilizers to agricultural land is a widespread agronomic practice but, although the importance of these practices has been noted in trace-element and heavy-metal cycling [e.g. 44], their possible influence on cycling of long-lived radionuclides does not appear to have been studied.

2.1.2 Mobility in soils

It has been stated [15] that "one of the greatest areas of uncertainty in assessing the long-term transfer of activity to foodstuffs is the predictions of the migration of radionuclides down through soil and of any physical or biochemical processes that modify their availability for uptake into plants with time". In this context it is useful to refer to the discussion of Loneragan [48] who stated that the concentrations of trace elements in soil solution are generally so low that the total quantity present at any one time would sustain relatively little plant growth. To provide a sufficient supply for growth the trace elements require rapid replenishment from the solid phase. Fuller [28] recorded the following factors as having a significant effect on the mobility of hazardous metals in soils:

- Soil texture or particle size distribution.
- Pore space distribution.
- Content and distribution of iron, aluminium and manganese hydroxy oxides in soil and coating particles.
- pH of soil and buffering capacity.
- Redox potential in soil in micro- as well as macro-pores.
- Soil organic matter and amount and concentration of organic constituents in wastes.

- Concentration of hazardous ion.

A general description of both the composition of soils and of the terrestrial geochemistry of the elements was given by Bowen [9], while a relevant discussion of the modelling of heavy metal behaviour in soils was provided by Harmsen [34] and the movement of ions in soils by Nye and Tinker [58]. The behaviour of radionuclides in soils was discussed by Scott-Russell [66] and more recently by Schwarzer [64, 65]. Schwarzer noted five processes which he considered to be most important in affecting the transport of radionuclides in soils. These were:

- sorption within the soil;
- hydrodynamic suspension;
- molecular diffusion;
- ground-water flow proper (convection);
- radioactive decay.

Bunzl [13] discussed and described models for several aspects of the transport and accumulation of radionuclides in the soil. These aspects included transport by diffusion and convection, sorption by solid-phase particles and the migration of radionuclides through soil containing channels. Baes [in:39] discussed a coefficient for soil-loss of radionuclides due to leaching (λ_{sl}) which was defined as:

$$\lambda_{sl} = \frac{V_w}{d_s[1+(\frac{\rho k_d}{\theta})]}$$

where

V_w = velocity of vertical water percolation
d_s = depth of soil root zone (cm)
ρ = soil bulk density (g cm^{-3})
θ = soil water content (ml cm^{-3})
k_d = equilibrium distribution coefficient of radionuclide species between soil and water (ml g^{-1}).

When $k_d = 0$ migration will occur with the soil water and as $k_d \to \infty$ no migration will occur. However, because the estimated range of λ_{sl} based on observed data included 3 to 5 orders of magnitude, Baes proposed that site-specific measurements of relevant parameters were required.

For the purpose of modelling radionuclide migration in soil the CEA/NRPB [15] categorised agricultural land into two types. The first 'undisturbed' type was used to simulate permanent pastures and the second 'well-mixed' type was used to simulate land that was subject to frequent ploughing or cultivation. The undisturbed soil model consisted of four compartments representing successive layers of soil of increasing depths. The contaminant was assumed to be well-mixed in each compartment. Resuspension and soil grazing were considered to occur from the first compartment, representing soil of 0 to 1 cm depth. The root zone of pasture plants was considered to extend to 15 cm depth and only material present in the fourth compartment (15 to 30 cm depth) was considered to be unavailable for uptake by plants. All transfer coefficients were based on the calculations of Simmonds et al. [69] which were from data for the migration of plutonium in soil. The coefficients were considered to be applicable to all other elements. Simmonds et al. referred to a transfer coefficient for migration of radionuclides from the root zone of pasture soil as 50 years but also stated that "the speculative nature of this estimation must however be recognised". Frissel and Jakubick [27] recently reviewed data for the transport of certain radionuclides in soil and observed a residence time for Cs-137 in soil of 2.5 y cm^{-1} with similar values for plutonium. These authors noted the wide range in observed and predicted residence times.

The CEA/NRPB well-mixed soil model consisted of one compartment representing soil of 0 to 30 cm depth in which a nuclide was assumed to be uniformly mixed and equally 'available'. Loss from the compartment was assumed to occur via downward processes in which diffusion and transport with general water movement were considered most important. The rate of loss was thus determined by the use of a single transfer coefficient.

Two recent studies are relevant to this well-mixed model. Crites et al. [21] provided data for the effects of ploughing on Am-241 distribution in an Enewetak soil. The fraction of Am-241 remaining in the surface soil after ploughing ranged from <5 to 25% of the initial amount with an average of 14%. Some surface activity was ploughed below the surface without mixing and represented from 25 to 50% of the activity of the surface layers. Horton et al. [40] provided data for the effect of cultivation on plutonium distribution in soil. In the twenty years before cultivation, the downward movement had been very slow. In discussion these authors stated that "tillage did produce slight increases in plutonium concentrations in the 5 to 15 cm depth of soil in both fields indicating some mixing of the 0 to 5 cm and 5 to 15 cm depths, but the amount of mixing was surprisingly small." Moreover in their conclusions they stated that "standard agricultural practices used in the S.E. United States will not greatly modify the distribution of plutonium in the soil and, therefore, will have relatively minor effects upon uptake by crop species."

The concept of a well-mixed soil model should, therefore, be considered with care. The effect of normal agricultural practices on elemental distribution in soil is not well documented and the assumption of an activity averaged throughout soil depths of 0 to 30 cm may underestimate the quantity available for uptake by plants. This factor is highly dependent on the rooting depth of the plant species concerned as well as the soil type and condition.

2.1.3 Loss from soils

Loss of activity from soil can occur via three main routes; resuspension can take place at the surface layer, plant uptake can occur at the surface or in intermediate layers, loss to groundwater via leaching can take place from the bottom layer. A further loss could also be expected to occur via lateral movement in soil but this component of loss is not well documented. Loss via leaching has been discussed in the context of the mobility of radionuclides in soil (Section 2.1.2).

Resuspension is a process whereby deposited activity may be removed from the surface layers of a soil, either in its original form or in association with soil particles. In some methodologies this process has been modelled on the basis of saltation rates [e.g. 13]. More commonly, a time-dependent resuspension factor has been used, but the initial value of this factor is recognised as being uncertain. In one study [15], the resuspension function, K_t, used was:

$$K_t = 10^{-5} \exp(-[\lambda_1 + \lambda_2 + \lambda]t) + 10^{-9} \exp(-[\lambda_2 + \lambda]t) \quad m^{-1}$$

where

t = time after initial deposition (s)
λ_1 = decay constant for initial decline (s^{-1})
λ_2 = decay constant for longer-term decline (s^{-1})
λ = radioactive decay constant of nuclide of interest.

Values of λ_1 of $1.46x10^{-7}$ s^{-1} and λ_2 of $2.2x10^{-10}$ s^{-1} were chosen, representing half lives of 0.15 and 100 years respectively. The data were considered insufficient to warrant a distinction between resuspension from undisturbed compared to ploughed land and the resuspension model was considered to be equally applicable to both rural and urban areas. It was also assumed that the same resuspension factor could be applied to activity deposited under either dry or wet conditions, although the function used was noted to overestimate the significance of the latter. Slinn [70] discussed the concept of the resuspension factor and values for the parameter. He noted the large variability (11 orders of magnitude) in tabulated resuspension factors and Travis [72] has noted the shortcomings of the definition of resuspension factors. Linsley [45] reviewed data for the resuspension of transuranic elements and proposed that, for a few weeks after their deposition, a resuspension factor of 10^{-6} m^{-1} could normally be applied. Only in conditions of moderate activity (defined as regular disturbance by vehicles or pedestrians) was an initial value of the resuspension factor of 10^{-5} m^{-1} considered to be appropriate. Recent experimental data for the resuspension of particulate matter from both grass and soil were provided by Garland [29]. The resuspension factors determined had initial values of $2x10^{-7}$ to 10^{-5} m^{-1} and declined as the reciprocal of time and increased as the square or cube of wind speed. Interestingly, a large fraction of resuspended material was deposited again within 3 metres of the point of resuspension. Garland deduced an upper limit of $7x10^{-11}$ m^{-1} for a fifteen year old deposit and concluded that the fraction of deposited fallout resuspended over thirteen years would not much exceed 10% of the deposit. Reynolds and Slinn [60] provided data for resuspension rates from various surfaces, these were greatest from gravel and least from soil or mown grass. The following average resuspension rates were reported: mown grass - $1.3x10^{-8}$ s^{-1}; soil - $1.3x10^{-8}$ s^{-1}; gravel - $5.6x10^{-8}$ s^{-1}. These authors also suggested that variations of resuspension with time were more the result of seasonal variations than the result of depletion by fixation to non-resuspendable materials. Healey [35] has recently provided a review of the various resuspension models that are available and recommended that mass-loading approaches should be used in generic studies; in this respect, Sehmel [66] concluded that there are more theoretical resuspension models available than data to validate or to use in those models.

Loss from soil via plant uptake will be determined by a wide range of factors. In some methodologies an estimate can be obtained by application of the plant:soil transfer coefficient (see Section 3.2) and the productivity of the agricultural system studied. This is not often done. Factors that are involved in soil-plant interactions have been discussed by Lonergan [48] and Nishita et al. [57]; while more recent studies have been reviewed and discussed by Nye and Tinker [58] and Tinker [71]. The proximity, extent and pattern of contact between soil and root are important in the absorption of ions from soils, particularly for those ions which are tightly bonded to soil colloids. The pattern of distribution of roots is influenced by both chemical and physical properties of soils. The uptake of a contaminant will be affected by the biological activity of the soil, while the interaction of plants and soils can change the composition of the soil solution or the root surface. The plant root can also modify the soil environment in its immediate vicinity both by excretion of chemically active substances and by absorption of water and ions. Plant exudates can influence the solubility (and hence the transport) of elements in soil solution as well as stimulating or reducing microbial activity. Further discussion of the interaction of plants with soils and their effect on the loss of nuclides from soils is given in the following sections of the report. However, the comments of Scott-Russell [66] that "no general principles can be laid down as to the manner in which measurements of absorption from the soil are most appropriately expressed. The choice of procedure should depend on the conditions of observation and on the purpose for which the results are to be used", are very relevant.

2.2 Plants

2.2.1 Input to plants

Plants can absorb radionuclides either in their above-ground parts by foliar absorption or in their below-ground parts by root absorption. In the preparation of models for plants it is advisable to consider the above-ground parts of the plants as separate external and internal components. This allows account to be taken of the surface contamination of vegetation by deposited particles. Nuclides can be deposited on the external plant component directly from the atmosphere by wet or dry deposition, via resuspension from soil and via contamination from either aspersion or irrigation waters.

(a) Dry deposition to external plant parts

Dry deposition to plants, as to soils, can be estimated by the concept of deposition velocity (V_g, discussed in Section 2.1.1). The derivation of, and background to, deposition velocity is pertinent in understanding the processes involved in this route of accumulation of radionuclides by plants. Knowledge of this subject is, to a large extent, the result of study by Chamberlain and co-workers and relevant data were summarised by Little [47]. Little remarked that the following three methods could account for particle capture by natural surfaces:

- Gravitational sedimentation.
- Inertial impaction.
- Eddy diffusion deposition.

The extent of particle capture will be determined by the physical characteristics of the particles, their concentration in air and the ambient meteorological conditions. Little also summarised considerable experimental data for deposition velocities of various particulates to vegetative surfaces. For exhaust lead particles labelled with Pb-203 these ranged from an upper value of 2.35×10^{-2} m s^{-1} for 0.02 μm diameter particles deposited on beech petioles to a lower value of 8×10^{-5} m s^{-1} for 0.2 μm diameter particles depositing on beech laminas. For polystyrene spheres, V_g ranged from an upper value of 2.2×10^{-1} m s^{-1} for 8.5 μm diameter particles depositing on nettle stems at a wind speed of 2.5 m s^{-1} to a lower value of 2.7×10^{-4} m s^{-1} for 2.8 μm diameter particles depositing on beech leaves also at a wind speed of 2.5 m s^{-1}. Deposition velocities to vegetation are closely and complexly related to particle size. A minimal deposition to grass at a wind speed of 2.5 m s^{-1} can be expected to occur with 0.3 μm diameter particles having a V_g of about 2.5×10^{-4} m s^{-1}. However, the actual V_g will also depend to a large extent on the physical characteristics of the plant crop being studied. As a result it is difficult to incorporate an overall V_g in either the discussion or modelling of deposition onto plants hence, in the latter case, the concept of the 'interception factor' has been used (see below). The CEA/NRPB [15] used a value for V_g of 5×10^{-3} m s^{-1} for all radionuclides studied apart from the noble gases and iodine. Bayer [4] used a value of 3×10^{-3} m s^{-1} for all radioisotopes other than iodine and the noble gases, and a value of 1×10^{-2} m s^{-1} for iodine isotopes. Heinemann and Vogt's studies [37] of the deposition of iodine agree with the value chosen by Bayer, in that the deposition velocity to grass averaged over the whole pasture time was 2×10^{-2} m s^{-1}. The measured value for aerosols was a factor of twenty times less than that of elemental iodine (i.e. 10^{-3} to 1×10^{-5} m s^{-1}) while laboratory measurements on methyl iodide produced a value which was only about 0.5% of that of elemental iodine; this value is similar to that of 5×10^{-5} m s^{-1} assumed by CEA/NRPB for organic forms of iodine. Moore et al. [54] recommended the following deposition velocities for vegetative surfaces:

- 2×10^{-2} m s^{-1} for reactive gases;
- 1×10^{-3} m s^{-1} for small particles (<4 μm diameter);
- 1×10^{-4} m s^{-1} for relatively unreactive gases.

(b) The interception factor

The interception factor (r) is the proportion of the total deposition (wet + dry) that is retained by the vegetation under study. Eriksson [25] discussed extensive experimental data concerning the interception of ionic or particulate radionuclides by pasture grasses and demonstrated that interception and retention were affected by humidity, precipitation and the form of material applied. The relative amount intercepted was reported to decrease in the following order: wet-deposited radionuclides > wet-deposited particles > particles dry-deposited on grass wet with rain > particles dry-deposited on grass superficially wet > particles dry-deposited on dry grass, and small particles > larger particles. Witherspoon and Taylor [77] reported data for the interception of particles by sorghum and soyabeans and showed a similar overall interception. Their data and other experimental data were included in the papers of Miller [51, 52] in which the model proposed by Chamberlain was validated. Chamberlain had provided a model to relate r to the biomass of the vegetation such that: